



Europäisches Patentamt
European Patent Office
Office européen des brevets



⑪ Publication number:

0 497 012 A1

⑫

EUROPEAN PATENT APPLICATION

⑬ Application number: 91202910.5

⑭ Int. Cl. 5: C08L 71/12, C08L 25/06,
C08K 3/34

⑬ Date of filing: 11.11.91

⑬ Priority: 29.01.91 NL 9100149

⑬ Applicant: GENERAL ELECTRIC COMPANY
1 River Road
Schenectady, NY 12345(US)

⑬ Date of publication of application:
05.08.92 Bulletin 92/32

⑬ Inventor: Ketels, Hendrikus Hubertus
Theodoor Maria
General Electric Plastics B.V., P.O. Box 117
NL-4600 AC Bergen op Zoom(NL)
Inventor: Di Fede, Cinzia Aurora Rita
General Electric Plastics B.V., P.O. Box 117
NL-4600 AC Bergen op Zoom(NL)
Inventor: Savenije, Hermannus Bernardus
General Electric Plastics B.V., P.O. Box 117
NL-4600 AC Bergen op Zoom(NL)

⑬ Designated Contracting States:
DE ES FR GB IT NL

⑬ Representative: Pratt, Richard Wilson et al
London Patent Operation G.E. Technical
Services Co. Inc. Essex House 12/13 Essex
Street
London WC2R 3AA(GB)

⑮ Polymer mixture based on polyphenylene ether and talcum.

⑯ The invention relates to a polymer mixture based on a polyphenylene ether, a polystyrene and/or a high-impact polystyrene, an agent to improve the impact strength, and talcum. By using a mixture of two linear block copolymers as agent (C), a polymer mixture is obtained having a favourable combination of low gloss, good impact strength and a good flow in the melt.

EP 0 497 012 A1

The invention relates to a polymer mixture based on a polyphenylene ether, polystyrene and/or high-impact polystyrene, an agent to improve the impact strength, and talcum, and to articles formed from the polymer mixture.

Such mixtures are described in US-A-4,483,958. The known polymer mixtures comprise a polyphenylene ether, a high impact polystyrene, a thermoplastic elastomer and an inorganic filler. The filler may be talcum. These known polymer mixtures have a high gloss and a good impact strength according to Izod, due to the use of a weight ratio between the quantity of inorganic filler and thermoplastic polymer of more than two. It appears from a comparative example of this Patent Specification that a lower gloss is obtained when a weight ratio of 4:6 (comparative example 6) is used.

10 The invention provides polymer mixtures which comprise the constituents described hereinbefore and which have a good impact strength, an as low as possible gloss and good flow properties of the polymer melt.

The polymer mixture according to the invention is characterised in that the polymer mixture is composed as follows:

- 15 A. 20-60 parts by weight of a polyphenylene ether,
- B. 40-70 parts by weight of a polystyrene or a high-impact polystyrene,
- C. 5-20 parts by weight of an agent to improve the impact strength,
- D. 2-10 parts by weight of talcum, the agent (C) being a mixture of two linear block copolymers.

It has been found that the desired combination of properties is difficult to obtain or cannot be obtained 20 when inorganic fillers other than talcum are used. By combination of a mixture of two agents to improve the impact strength it has been found possible to obtain a good combination of impact strength, flow properties and low gloss.

The agent to improve the impact strength (constituent C) preferably comprises:

- C1. 35-65% by weight of a linear A-B1-A' block copolymer
- 25 and
- C2. 35-65% by weight of a linear A"-B2-A'" block copolymer, wherein
- A, A', A" and A''' are equal or different terminal blocks which are derived from a vinylaromatic compound and have a weight-averaged molecular weight from 2,000 to 100,000;
- B1 is a block derived from a conjugated diene monomer,
- 30 and
- B2 is a block derived from one or more alkylene monomers, the blocks B1 and B2 having a weight-averaged molecular weight from 10,000 to 500,000.

The polymer mixture is preferably composed so as to comprise 20-35 parts by weight of constituent A, 45-65 parts by weight of constituent B, 10-15 parts by weight of constituent C and 4-8 parts by weight of constituent D.

The polymer mixture according to the invention comprises at any rate one or more of the following constituents:

- A. a polyphenylene ether;
- B. a polystyrene and/or a high-impact polystyrene;
- 40 C. a mixture of at least two block copolymers as defined in the Claims;
- D. talcum.

In addition to the constituents mentioned hereinbefore the polymer mixture may comprise one or more of the additives conventionally used for polymer mixtures, for example, stabilisers, agents to improve the flame-retarding properties, dyes, pigments and plasticisers.

45 The polymer mixture according to the invention can be obtained according to the conventional techniques for preparing polymer mixtures. The polymer mixture according to the invention is preferably manufactured by compounding.

The polymer mixture according to the invention may be formed according to the conventional techniques for processing synthetic resins to articles, for example, by extrusion or by injection moulding.

50 The nature of the constituents to be used will be described in greater detail hereinafter.

A. POLYPHENYLENE ETHER

55 Polyphenylene ethers are generally known polymers. All polyphenylene ethers known per se may be used in the polymer mixtures according to the invention. An elaborate enumeration of suitable polyphenylene ethers is to be found, for example, in WO 87 / 00540.

Particularly suitable are poly(2,6-dimethyl-1,4-phenylene)ether, poly(2,6-diethyl-1,4-phenylene)ether, poly(2-methyl-6-ethyl-1,4-phenylene)ether. It is also possible to use copolymers, for example, copolymers

which comprise units derived from 2,3,6-trimethyl phenol and from 2,6-dimethyl phenol.

B. POLYSTYRENE AND/OR HIGH-IMPACT POLYSTYRENE

5 High-impact polystyrene consists of a mixture of a rubber and a polystyrene prepared from a vinylaromatic compound, for example, styrene itself and/or alpha-methyl styrene and/or dimethyl styrene compounds. The rubber usually consists of a conjugated diolefins polymer, for example, polybutadiene. Other suitable rubbers are EPDM and acrylate rubbers. It is also possible to use a polystyrene without rubber.

10 C. AGENT TO IMPROVE THE IMPACT STRENGTH

The polymer mixture according to the invention comprises a mixture of at least two linear block copolymers as an agent to improve the impact strength. The mixture preferably comprises a mixture of:

15 C1. 35-65% by weight of a linear A-B1-A' block copolymer
and
C2. 35-65% by weight of a linear A"-B2'A''' block copolymer, wherein
A, A', A" and A''' are equal or different terminal blocks which are derived from a vinylaromatic compound
20 and have a weight-averaged molecular weight from 2,000 to 100,000; B1 is a block derived from a conjugated diene monomer,
and
B2 is a block derived from one or more alkylene monomers, the blocks B1 and B2 having a weight-averaged molecular weight from 10,000 to 500,000.

25 The agent even more preferably comprises a mixture of 40-60% by weight of constituent C1 and 60-40% by weight of constituent C2.

As stated hereinbefore, the terminal blocks A, A', A" and A''' are derived from vinylaromatic compounds. Suitable vinylaromatic compounds are styrene, alpha-methyl styrene, vinyl toluene, vinyl xylene and vinyl naphthalene. A, A', A" and A''' may be equal or different.

30 C. TALCUM

The polymer mixture according to the invention comprises talcum. The talcum to be used preferably has a particle size between 1.5 and 10 micrometres. Particles smaller than 1.5 micrometres result in a polymer mixture having less favourable properties. Talcum particles having a size of more than 10 micrometres result in a deterioration of the impact strength according to Izod.

35 The invention will now be described in greater detail with reference to the ensuing specific examples.

EXAMPLES

40 The following constituents were used in the examples below:

PPE: a polyphenylene-ether derived from 2,6-dimethylphenol having an intrinsic viscosity of 38 ml/g measured at 250 C in chloroform.

HIPS: a high-impact polystyrene having a butadiene rubber; rubber content approximately 10% by weight.

I.M. 1: a linear block copolymer A-B1-A', wherein A and A' are polystyrene blocks and B1 is a polybutadiene block, having a weight ratio of the total of the styrene blocks to the polybutadiene block of approximately 30 to 70 and a number-averaged molecular weight of the block copolymer of approximately 60,000.

I.M. 2: a linear block copolymer A"-B2-A''', wherein A" and A''' are polystyrene blocks having a number-averaged molecular weight of 16,000 and B2 is an ethylene-butylene copolymer having a number-averaged molecular weight of 78,000, with a weight ratio of the styrene blocks to ethylene-butylene blocks of 30/70 and an average molecular weight by number of the blockcopolymer of about 110,000.

Talcum: a talcum product having an average particle size of 4.5 micrometres.

Stab: a mixture of conventionally used stabilisers.

C.B.: carbon black

PE: polyethylene

The constituents mentioned hereinbefore were compounded in an extruder in the quantities recorded in the table hereinafter. The average temperature adjustment of the extruder was 280 C.

Standardised test pieces were manufactured from the polymer mixtures thus obtained in an injection

moulding machine. The test pieces were subjected to the following tests. The impact strength was determined according to Izod (ASTM 256) and according to DIN 53443 (Falling Dart Impact) at room temperature and at minus 30 °C. The gloss was determined according to ASTM D523 at an angle of 60° and of 85°. The melt viscosity was measured according to ASTM D3835 at a shearing rate of 1,500 per second. The found results are also recorded in the table hereinafter.

TABLE

	Example	A*	B*	C*	I
10	Composition				
	(parts by weight)				
15	o PPE	27.9	27.9	27.9	27.9
	o HIPS	54.6	54.6	54.6	54.6
15	o I.M. 1	--	--	13.5	6.75
	o I.M.2	13.5	13.5	--	6.75
20	o Talcum	--	6.0	6.0	6.0
	o Stab	0.6	0.6	0.6	0.6
20	o C.B.	1.9	1.9	1.9	1.9
	o PE	1.5	1.5	1.5	1.5
25	* comparative examples				
	Properties				
30	o Izod impact strength (J/M)				
	23 °C	458	280	331	305
	-30 °C	170	88	167	121
35	o Falling Dart Impact (J)				
	23 °C	52	65	65	71
	-30 °C	80	55	58	49
40	o Gloss				
	60°	25	15	7	11
	85°	80	65	46	57
45	o Melt viscosity				
	(Pa.s)	170	176	219	184
	at 1500 s ⁻¹				

50 It may be seen from the results as recorded in the table that the polymer mixture according to the invention (Example I) has an optimum combination of impact strength, low gloss and melt viscosity. Comparative example A has the best impact strength according to Izod and the best flow properties, but for many applications its gloss is too high. Addition of talcum to the composition according to comparative example A results in a decrease of the gloss, but also in a substantial reduction of the impact strength at room temperature and at lower temperatures (see comparative example B). Replacing the agent to improve the impact strength (i.e. I.M.1) by another agent (i.e. I.M.2) results in an improvement of the impact strength, in a further reduction of the gloss, but in too high an increase of the melt viscosity (see comparative

example C). By using a mixture of I.M. 1 and I.M. 2 (example I according to the invention) a good combination of impact strength, low gloss and melt viscosity is obtained, with a remarkably good Falling Dart impact strength.

5 All the patent publications mentioned hereinbefore are considered to be incorporated in this text by reference.

Claims

- 10 1. A polymer mixture based on a polyphenylene ether, polystyrene and/or high-impact polystyrene, an agent to improve the impact strength, and talcum, characterised in that the polymer mixture has the following composition:
 - A. 20-60 parts by weight of a polyphenylene ether,
 - B. 40-70 parts by weight of a polystyrene and/or a high-impact polystyrene,
 - C. 5-20 parts by weight of an agent to improve the impact strength,
 - 15 D. 2-10 parts by weight of talcum, the agent (C) comprising a mixture of two linear block copolymers.
- 20 2. A polymer mixture as claimed in Claim 1, characterised in that constituent C comprises:
 - C1. 35-65% by weight of a linear A-B1-A' block copolymer and
 - C2. 35-65% by weight of a linear A"-B2-A''' block copolymer, wherein A, A', A" and A''' are equal or different terminal blocks derived from a vinylaromatic compound and have a weight-averaged molecular weight from 2,000 to 100,000;
 - 25 B1 is a block derived from a conjugated diene monomer, and
 - B2 is a block derived from one or more alkylene monomers, the blocks B1 and B2 having a weight-averaged molecular weight from 10,000 to 500,000.
- 30 3. A polymer mixture as claimed in Claim 1, characterised in that the polymer mixture comprises 20-35 parts by weight of constituent A, 45-65 parts by weight of constituent B, 10-15 parts by weight of constituent C and 4-8 parts by weight of constituent D.
4. Articles formed from the polymer mixture as claimed in Claim 1.

35

40

45

50

55



European Patent
Office

EUROPEAN SEARCH REPORT

Application Number

EP 91 20 2910

DOCUMENTS CONSIDERED TO BE RELEVANT			
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int. CL.5)
D, Y	US-A-4 483 958 (NABUTOSHI KOSAKA ET AL.) * claims *	1-4	C08L71/12 C08L25/06 C08K3/34
Y	US-A-4 277 575 (WILLIAM H. HAAF ET AL.) * claims; examples *	1-4	
Y	FR-A-2 375 297 (GENERAL ELECTRIC COMPANY) * page 4, line 22 - line 33; claims; examples 3,6,9 *	1-4	
A	US-A-4 252 913 (ARTHUR KATCHMAN ET AL.) * claims; example 7 *	1	
The present search report has been drawn up for all claims			
Place of search	Date of compilation of the search	Examiner	
THE HAGUE	07 MAY 1992	DE LOS ARDOS E.	
CATEGORY OF CITED DOCUMENTS			
X : particularly relevant if taken alone	T : theory or principle underlying the invention		
Y : particularly relevant if combined with another document of the same category	E : earlier patent document, but published on, or after the filing date		
A : technological background	D : document cited in the application		
O : non-written disclosure	L : document cited for other reasons		
P : intermediate document	A : number of the same patent family, corresponding document		

BEST AVAILABLE COPY